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6. AUTHOR(S) Dr Shaul Mukamel				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Dept of Chemistry University of Rochester River Campus, Hutchison Hall Rochester NY 14627-0216			8. PERFORMING ORGANIZATION REPORT NUMBER AFOS R-TR- 96-0055	
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13. ABSTRACT (Maximum 200 words)
A microscopic theory for the nonlinear optical response and susceptibilities of molecular materials with localized and delocalized electronic states (e.g. molecular assemblies, conjugated polymers, aggregates and monolayers) was developed. The theory, which properly accounts for many-body effects, is based on the derivation of coupled reduced equations of motion for the material variables, which determine the optical response. The present formulation maps the calculation of optical nonlinearities onto solving the dynamics of coupled nonlinear oscillators and overcomes the difficulties associated with the local-field approximation. An operational definition of the nonlinear coherence-size N_c , which controls the cooperative enhancement of the optical response is developed. A real-space representation of the optical response of conjugated polyenes is developed by using the Wannier representation to derive equations of motion for coupled two-site oscillators representing correlated electron-hole pairs. The resulting elementary excitations are shown to be intermediate between the molecular (Frenkel) and the semiconductor (Wannier) excitons, and clearly resemble charge transfer excitons. Theoretical analysis of resonant and off-resonant femtosecond pump-probe absorption experiments on polydiacetylene-para-toluene sulfonate (PTS) was carried out using a 3-mode Brownian oscillator model for the nuclear dynamics. The role of spectral diffusion

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Molecular Nonlinear Optical Susceptibilities in Condensed Phases Final Technical Report

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Summary

A microscopic theory for the nonlinear optical response and susceptibilities of molecular materials with localized and delocalized electronic states (e.g. molecular assemblies, conjugated polymers, aggregates and monolayers) was developed. The theory, which properly accounts for many-body effects, is based on the derivation of coupled reduced equations of motion for the material variables, which determine the optical response. The present formulation maps the calculation of optical nonlinearities onto solving the dynamics of coupled nonlinear oscillators and overcomes the difficulties associated with the local-field approximation. An operational definition of the nonlinear coherence-size N_c , which controls the cooperative enhancement of the optical response is developed. A real-space representation of the optical response of conjugated polyenes is developed by using the Wannier representation to derive equations of motion for coupled two-site oscillators representing correlated electron-hole pairs. The resulting elementary excitations are shown to be intermediate between the molecular (Frenkel) and the semiconductor (Wannier) excitons, and clearly resemble charge transfer excitons. Theoretical analysis of resonant and off-resonant femtosecond pump-probe absorption experiments on polydiacetylene-para-toluene sulfonate (PTS) was carried out using a 3-mode Brownian oscillator model for the nuclear dynamics. The role of spectral diffusion process was explored.

This grant resulted in 46 publications (which are listed below). In addition, a graduate level textbook entitled "Nonlinear Optical Spectroscopy: The Density Matrix Perspective" by

S. Mukamel is being completed. The book develops a unified theoretical microscopic treatment of the optical nonlinear response of molecular, conjugated-polymer, and semiconductor materials.

Publications Resulting from the AFOSR Award

"Nonlinear Susceptibilities of Donor-Acceptor Conjugated Systems; Coupled-Oscillator Representation," G. Chen and S. Mukamel, J. Am. Chem. Soc. **117**, 4945 (1995).

"Two Exciton Collective Photon Echoes in Disordered Molecular Nanostructures," V. Chernyak and S. Mukamel, Phys. Rev. Lett. **74**, 4895-4898 (1995).

"Four Wave Mixing and Fluorescence of Confined Excitons in Molecular Aggregates and Nanostructures," V. Chernyak, N. Wang, and S. Mukamel, Physics Reports (in press)

"Time and Frequency Resolved Luminescence from Excitonic Nanostructures," by V. Chernyak and S. Mukamel, Physica Status Solidi **188**, 275-284 (1995).

"Superexchange Versus Sequential Long Rang Electron Transfer; Density Matrix Pathways in Liouville Space," S. Skourtis and S. Mukamel, Chem. Physics **197**, 367-388 (1995).

"Energy Transfer, Spectral Diffusion, and Fluorescence of Molecular Aggregates: Brownian Oscillator Analysis," S. Mukamel and V. Rupasov, Chem. Phys. Lett. **242**, 17-26 (1995).

"Gauge Invariant Formulation of Molecular Electrodynamics and the Multipolar Hamiltonian," V. Chernyak and S. Mukamel, Chem. Phys. **198**, 133-143 (1995).

"Dominant Electronic Oscillators in the Optical Nonlinearities of Conjugated Polyenes," G. Chen and S. Mukamel, Chem. Phys. Lett. **240**, 296-303 (1995).

"High-Order Echoes in Vibrational Spectroscopy of Liquids," V. Khidekel and S. Mukamel, Chem. Phys. Lett. **240**, 304-314 (1995).

"Semiclassical Theory of Molecular Nonlinear Optical Polarization," M. A. Sepúlveda and S. Mukamel, J. Chem. Phys. **102**, 9327-9344 (1995).

"Femtosecond Pump-Probe Spectroscopy of Intermolecular Vibrations in Molecular Dimers," Y. Tanimura and S. Mukamel, J. Chem. Phys., **103**, 1981-1994 (1995).

"Multi-State Quantum Fokker-Planck Approach to Nonadiabatic Wavepacket Dynamics in Pump-Probe Spectroscopy," Y. Tanimura and S. Mukamel, J. Chem. Phys. **101**, 3049-3061 (1994).

"Electronic Coherence and Nonlinear Susceptibilities of Conjugated Polyenes," S. Mukamel, A. Takahashi, H. X. Wang, and G. Chen, Science **266**, 250 - 254 (1994).

"Collective Charge Density Fluctuations and Nonlinear Optical Response of C₆₀", A. Takahashi, H. X. Wang, and S. Mukamel, Chem. Phys. Lett., **216**, 394-398 (December, 1993).

"Temperature Dependence and Non-Condon Effects in Pump-Probe Spectroscopy in the Condensed Phase", Y. Tanimura and S. Mukamel, J. Opt. Soc. Am. B, **10**, 2263-2268 (December, 1993).

"Self Broadening and Exciton Line Shifts in Gases: Beyond the Local Field Approximation", J. A. Leegwater and S. Mukamel, Phys. Rev. A, **49**, 146-155 (January, 1993).

"The Optical Dielectric Function of Polarizable Liquids", J. A. Leegwater and S. Mukamel, J. Chem. Phys. **99**, 6062-6070 (October, 1993).

"Four Wave Mixing Signatures of Exciton Bose Condensation", J. A. Leegwater and S. Mukamel, Chem. Phys. Lett., **217**, 456-460 (January, 1994).

"Two-Dimensional Femtosecond Vibrational Spectroscopy of Liquids", Y. Tanimura and S. Mukamel, J. Chem. Phys., **99**, 9496-9511 (December, 1993).

"Mode-Locking Matter with Light," B. Kohler, J. L. Krause, F. Raksi, C. Rose-Petruck, R. M. Whitnell, K. R. Wilson, V. V. Yakovlev, Y. Yan and S. Mukamel, J. Phys. Chem., **97**, 12602-12608 (December, 1993)

"Anharmonic Oscillator Modeling of Nonlinear Susceptibilities and its Application to Conjugated Polymers," A. Takahashi and S. Mukamel, J. Chem. Phys., **100**, 2366-2384 (February, 1994)

"Cooperative Ultrafast Nonlinear Optical Response of Molecular Nanostructures," N. Wang, V. Chernyak, and S. Mukamel, J. Chem. Phys., **100**, 2465-2480 (February, 1994).

"Exciton Confinement and Nonlocal Nonlinear Optical Response of Organic Multiple Quantum Wells," N. Wang, J. K. Jenkins, V. Chernyak, and S. Mukamel, Phys. Rev. B **49**, 17079 (June, 1994).

"Multi-State Quantum Fokker-Planck Approach to Nonadiabatic Wavepacket Dynamics in Pump-Probe Spectroscopy," Y. Tanimura and S. Mukamel, J. Chem. Phys. **101**, 3049-3061 (August, 1994).

"Simulation of the Ultrafast Optical Response of Water", L. E. Fried and S. Mukamel, in Time-Resolved Vibrational Spectroscopy V, H. Takahashi Editor, Springer-Verlag, Berlin, (1992) pp. 295- 298.

"Many-body Effects In Nonlinear Susceptibilities; Beyond the Local-field Approximation", S. Mukamel, in Nonlinear Optical Properties of Organic Molecules and Crystals, Volume 3, J. Zyss, Editor, Academic Press, New York (1993).

"Interplay of Excitonic and Phonon-Mediated Stark Effects in Quantum Wells", J. R. Kuklinski and S. Mukamel, J. Lum. **53**, 97-100, 1992.

"Brownian Oscillator Analysis of Femtosecond Pump-Probe Spectroscopy of Polydiacetylene", W.B. Bosma, S. Mukamel, B.I. Greene, and S. Schmitt-Rink, Synthetic Metals, **49**, 71-76 (1992).

"Size Scaling and Exciton Coherence-size in the Optical Nonlinearities of Conjugated Polyenes", S. Mukamel and H.X. Wang, in Optics of Semiconductor Nanostructures, F. Henneberger, S. Schmitt-Rink and E. O. Göbel, Editors, (Akademie Verlag, Berlin, 1993) p. 361-395.

"Quasiparticle Exciton Representation of Frequency Dispersed Optical Nonlinearities of Conjugated Polyenes", H.X. Wang and S. Mukamel, J. Chem Phys., **97**, 8019-8036, (1992).

"Exciton Transport and Degenerate Four Way of Mixing in Topologically Disordered Systems", N. Wang, J.A. Leegwater, S. Mukamel, J. Chem Phys. **98**, 5899 - 5911 (1993).

"Real-Time Path Integral Approach to Quantum Coherence and Dephasing in Nonadiabatic Transitions and Nonlinear Optical Response", Y. Tanimura and S. Mukamel, Phys. Rev. A., **46**, 118-136 (1992).

"Simulation of Nonlinear Electronic Spectroscopy in the Condensed Phase", L.E. Fried and S. Mukamel, Adv. Chem. Phys., **84**, 435-516 (1993).

"Quantum Electrodynamics of Molecular Nanostructures", J. Jenkins and S. Mukamel, J. Chem Phys., **98**, 7046-7058 (1993).

"Simulation of the Intermolecular Vibrational Spectra of Liquid Water and Water Clusters", W.B. Bosma, L.E. Fried, and S. Mukamel, J. Chem Phys. **98**, 4413-4421 (1993).

"Transient Grating Spectroscopy of Exciton Sound Waves in Dense Exciton Fluids", Jan A. Leegwater and Shaul Mukamel, Chem. Phys. Lett., **203**, 125-130 (1993).

"Quantum Brownian Oscillator Analysis of Pump-Probe Spectroscopy in the Condensed Phase", Yoshitaka Tanimura and Shaul Mukamel, in Ultrafast Spectroscopy in Chemical Systems, pp. , John D. Simon (Editor), (Kluwer Academic Publishers, 1993).

"Quantum Confinement and Nonlinear Optical Response of Conjugated Molecules", H. X. Wang, A. Takahashi, M. Hartmann and S. Mukamel, *SPIE OE/LASE 93, Los Angeles, January 16 - 23 (1993)*

"Femtosecond Four Wave Mixing Spectroscopy of Conjugated Polymers", M. Hartmann and S. Mukamel, J. Chem. Phys., **99**, 1597-1606 (1993)

List of Personnel involved in the AFSOR Sponsored Research

Research Associates:

- Dr. Guanhua Chen, Ph.D. in Physics, 1992, California Institute of Technology, California, USA
- Dr. Thomas Wagersreiter, Ph.D. in Physics, 1993, University of Wien, Austria (supported by the Schroedinger fellowship)
- Dr. Miguel Sepulveda, Ph.D. in Chemistry, 1992, University of Washington, USA.
- Dr. Vadim Khideckel, Ph.D. in Chemistry, 1994, Tel Aviv University, Israel.

Visiting Scientists:

- Dr. Vladimir Chernyak, Ph.D., USSR Academy of Sciences, 1983.
- Dr. Valery Rupasov, Ph.D., USSR Academy of Sciences, 1989.

Graduate Students Supported:

- Mr. Pavel Rott, Department of Physics, University of Rochester (First Year).
- Mr. Sergei Tretiak, Department of Chemistry, University of Rochester (First Year).

Undergraduate Students:

- Mr. Uri Avissar, Summer Student; Currently, Chemistry Major, Yale University.

In addition, we are continuing to collaborate with several former postdoctoral fellows who currently hold faculty positions.

Dr. Akira Takahashi, Ph.D. Kyoto University, 1989; Currently, Assistant Professor, Yamaguchi University, working on applications to doped polymers and the optical signatures of polarons and solitons.

Dr. Jan Adrian Leegwater, Ph.D. University of Utrecht, Netherlands, 1991; Currently, Assistant Professor, Vrije Universiteit in Amsterdam; and
Dr. Yoshitaka Tanimura, Ph.D. Keio University, Japan, 1989; Currently, Assistant Professor, Institute for Molecular Science, Japan, working on vibrational dynamics and spectroscopy.

Dr. Michael Hartmann, Ph.D. University of Humboldt, Berlin, Germany, 1991; Currently, Assistant Professor, Max-Planck Arbeitsgruppe Festkoerpertheorie, working on femtosecond coherent spectroscopy.

Dr. David Beljonne, Ph.D. University of Mons, 1994; From J. L. Bredas' group, will visit Rochester this summer for 2 months to establish a collaboration.

Letters of support from Seth Marder, George Stegeman, and Jean-Luc Bredas expressing interest in close interaction and collaboration are enclosed.

INTERACTIONS/TRANSITIONS

a. Participation/Presentation at Meetings, Conferences, Seminars, etc. INVITED PRESENTATIONS

University of Indiana, Department of Chemistry, "Femtosecond Optical Spectroscopy of Molecular Systems in Condensed Phases" Bloomington, Indiana, November 1992.

University of Pittsburgh, Department of Chemistry, "Nonlinear Femtosecond Spectroscopy of Molecular Systems in Condensed Phases", Pittsburgh, Pennsylvania, November 1992.

Joint Harvard - MIT Seminar, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons and Molecular Nanostructures", Cambridge, Massachusetts, December 1992.

SPIE International Symposium on Advanced Electronic and Optoelectronic Materials, "Nonlinear Optical Response of Conjugated Systems : Electron-Hole Anharmonic-Oscillator Picture," Los Angeles, California, January 1993.

ACS National Meeting, "Nonlinear Optical Response of Conjugated Polyenes," Denver, Colorado, March 1993.

CLEO/QELS Quantum Electronics and Laser Science Conference, "Third Order Nonlinear Spectra of a Polydiacetylene: The Quasi-Particle Exciton Representation", Baltimore, Maryland, May 1993.

French-German-Israeli Symposium on *Dynamical Processes in Condensed Molecular Systems*, "Cooperative Effects in the Nonlinear Optical Response of Confined Excitons," Garchy, France, May 1993.

Royal Netherlands Academy Colloquium on *Femtosecond Reaction Dynamics*, "Multimode Brownian Oscillator Analysis of Ultrafast Nonlinear Optical Response," Amsterdam, Netherlands, May 1993.

Sixth International Conference on *Time-Resolved Vibrational Spectroscopy*, "Two-Dimensional Off-Resonant Femtosecond Spectroscopy of Liquids", Berlin, Germany, May 1993.

Max-Planck-Gesellschaft zur Förderung der Wissenschaften, Arbeitsgruppe *Halbleitertechnik*, "Nonlinear Optical Response of Molecular and Semiconductor Nanostructures", Berlin, Germany, May 1993.

ETH, Laboratorium für Physikalische Chemie, "Optical Nonlinearities and Femtosecond Spectroscopy of Confined Excitons", Zürich, Switzerland, May 1993.

Canadian Society for Chemistry Conference, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons", Montreal, Canada, June 1993.

DPC '93, 9th International Conference on the *Dynamical Processes in Excited States of Solids*, "Retardation Effects in the Nonlinear Optical Response; Pump Probe Spectroscopy of Nanostructures," Cambridge, Massachusetts, August 1993.

Russian Academy of Sciences Institute of Spectroscopy XXVth Anniversary Conference on *Modern Problems of Spectroscopy*, "Nonlinear Optical Response of Confined Excitons in Molecular and Semiconductor Nanostructures," Troitsk, Moscow, October 1993.

Air Force Nonlinear Optical Polymer Program Review, "Anharmonic Oscillator Representation of Optical Nonlinearities of Conjugated Polyenes," Washington, DC, November 1993.

Phillipps-Universität Marburg, Workshop on *Optical and Opto-electronic Properties of Conjugated Polymers*, "Anharmonic Oscillator Quasiparticle Representation of Optical Nonlinearities of Conjugated Polymers," Marburg, Germany, November 1993.

100th Birthday Anniversary Conference on *Nonlinear Laser Spectroscopy of Organic Dyes in Condensed Phase*, in memorium of Professor Günther Scheibe, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons in Molecular Assemblies," Physics Center, Bad Honnef, Germany, November 1993.

Tel Aviv University, "Optical Nonlinearities of Confined Excitons in Conjugated Polyenes and Molecular Nanostructures," Tel Aviv, Israel, December, 1993

SPIE Symposium on *Organic, Metallo-organic and Polymeric Materials for Nonlinear Optical Applications*, "Nonlinear Optics of Conjugated Molecules," Los Angeles, California, January 1994.

American Chemical Society National Meeting Symposium on *Chemical Dynamics in the Gas Phase*, "Nonlinear Spectroscopy of Clusters," San Diego, California, March 1994.

Brown University, Chemistry Colloquium, "Cooperative and Ultrafast Radiative Dynamics of Confined Excitons," Providence, Rhode Island, April 1994.

Boston University, Chemistry Colloquium, "Electrodynamics of Conjugated Polyenes and Molecular Nanostructures," Boston, Massachusetts, April 1994.

SIAM Meeting on *Materials Science*, "Anharmonic Oscillator Modeling of Nonlinear Susceptibilities in Molecular Nanostructures and Conjugated Polyenes," Pittsburgh, Pennsylvania, April 1994.

Ultrafast Phenomena, Ninth International Meeting on "Two-Dimensional Off-Resonant Femtosecond Spectroscopy of Liquids," Dana Point, California, May 1994.

Discussion Leader at the Gordon Conference on Electronic Processes in Organic Materials, Andover, NH, July 1994.

American Chemical Society National Meeting Symposium on *Theoretical and Computational Modeling of NLO and Electronic Materials*, "Anharmonic Oscillator Modeling of Nonlinear Susceptibilities and its Application to Conjugated Polymers," Washington D.C., August 1994.

Gordon Conference on Electron Donor-Acceptor Interactions, "Nonlinear Optical Response and Long-Range Electron Transfer in Conjugated Polyenes", Newport, RI, August 1994.

XIVth International Conference on Raman Spectroscopy, "Recent Development in Raman Theory and Perspective of Applications," Kowloon, Hong Kong, August 1994.

The Hong Kong University of Science and Technology, Physics and Chemistry Colloquium, "Nonlinear Optical Response of Confined Excitons in Conjugated Polymers and Molecular Nanostructures," Kowloon, Hong Kong, August 1994.

Fourth International Workshop on Nonlinear Optics and Excitation Kinetics in Semiconductors, "Cooperative Nonlinear Optical Response in Semiconductor and Molecular Nanostructures," Berlin, Germany, November 1994.

Universite de Mons-Hainaut, Seminar on "Optical Nonlinearities of Confined Excitons in Conjugated Polyenes and Molecular Nanostructures," Mons, Belgium, November, 1994.

Vrije Universiteit, Seminar on "Ultrafast Optical Nonlinearities of Confined Excitons in Molecular Assemblies," Amsterdam, The Netherlands, November, 1994.

Seminar on "Cooperative Effects and Femtosecond Nonlinear Optical Spectroscopy of Conjugated Polyenes, Molecular Aggregates, and Liquids," Michigan State University, East Lansing, MI, February 1995.

Seminar on "Nonlinear Optical Response of Confined Excitons in Conjugates in Polyenes and Molecular Nanostructures," University Minnesota, Minneapolis, March 1995.

Seminar on "Cooperative Nonlinear Response of Conjugated Polyenes and Molecular Aggregates," Iowa State University of Science & Technology, Ames, IA, March 1995.

Austrian-Israeli-German Symposium on Dynamical Processes in Condensed Molecular Systems, "Optical Nonlinearities in Molecular Nanostructures and Conjugated Polyenes," Vienna, Austria, May 1995.

Workshop on Electronic Nanostructures, "Optical Nonlinearities in Molecular Nanostructures and Conjugated Polyenes," Aohrus, Denmark, May 1995.

TRVS (Time-Resolved Vibrational Spectroscopy) Meeting, "Photon Echoes as Collective Resonance in Multidimensional Vibrational Spectroscopy of Liquids," Santa Fe, NM, June 1995.

Symposium on Nonlinear Optics, "Nonlinear Optical Spectroscopy in Condensed Phases," Aalborg, Denmark, July 1995.

Femtochemistry: The Lausanne Conference, "Semiclassical Approach to Nonlinear Spectroscopy using the Density Matrix in Liouville space," Lausanne, Switzerland, September 1995.

XXth International Solvay Conference in Chemistry on Photochemistry: Chemical Reactions and their Control on the Femtosecond Time Scale, Université Libre de Bruxelles, November 1995.

Symposium on New Developments in Ultrafast Time-Resolved Vibrational Spectroscopy, Tokyo, Japan, December 1995.

4th Pacific Polymer Conference on Recent Developments in Vibrational Spectroscopy, Honolulu, Hawaii, December 1995.

b. Consultative and advisory functions - None.

c. Transitions. Cases where knowledge resulting from your effort is used, or will be used - None.

NEW DISCOVERIES, INVENTIONS, OR PATENT DISCLOSURES

None at this time.

HONORS/AWARDS

Elected Chair of the Gordon Conference on Electronic Processes in Organic Materials (August, 1994).

Chair of the Nominating Committee for the Offices of the Division of Chemical Physics (September, 1994).

International Advising Committee of the International Conference on Optical Properties of Nanostructures, Sendai, Miyagi Japan, September 1994.

Miller Fellowship at the Departments of Chemistry and Physics, University of California, Berkeley (1996)

Vice Chair of the Gordon Conference on Electronic Processes in Organic Materials, Andover, NJ (1996)

Elected as a Member of the International Advisory Committee of the EXCON '96 Conference

Visiting Professor at the Université Louis Pasteur, Strasbourg, France (1996)